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A. Studies of Stratospheric Particulates (Task 673-62-13-20)

B. Investigators and Institutions:
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C. Research Objectives:
The principal objective of this work is to quantify the importance of heterogeneous chemistry to ozone depletion in the polar regions. The ambient stratospheric sulfate layer, and especially clouds in the polar stratosphere may provide surfaces upon which reactions occur and they may sequester or remove materials from the stratosphere. Our goals are to theoretically simulate such heterogeneous processes so that we may better quantify their importance, and so that we can identify processes that need to be studied in the laboratory. A sophisticated computer model of polar stratospheric clouds has been developed and used to study the properties of ice clouds. The model has recently been extended to investigate nitric acid clouds and ice clouds as well as their interactions with stratospheric gases. The model is now being applied to interpret data collected during recent expeditions to the Antarctic and the Arctic. Some work has also been done to understand the properties of noctilucent clouds and their implications for the chemistry and dynamics of the upper stratosphere.

D. Progress and Results:

A sophisticated numerical model of cirrus cloud microphysics has been developed and simulations of water ice clouds in the Antarctic atmosphere have been conducted. One conclusion of this work is that the particle sizes, optical depths and vertical mass transfer rates of ice

clouds are very sensitive to the cooling rates at which these clouds form. We also concluded that nitric acid is not incorporated into ice clouds by physical processes such as nucleation or coagulation, but rather by vapor transfer processes. We have also developed a theory which allows us to relate laboratory measurements of heterogeneous reaction rates to the reactions which occur on particle surfaces. We have additionally developed a theory for the vapor pressures of nitric acid trihydrate which seems to explain most of the laboratory data. We have analyzed data obtained during the Antarctic Ozone expedition and shown that the polar clouds that are present much of the time are not composed of water ice, but are composed of condensed nitric acid. We have recently improved the cloud model by treating nitric acid, hydrochloric acid and other condensed phase materials as minor constituents within ice and nitric acid particles. We are currently adding a gas phase chemistry package to the cloud model. This work will allow us to study more precisely how the ice clouds interact with the chemistry in the polar night. We have found that the brightness of noctilucent clouds is very sensitive to the supply of water vapor. Therefore, we have suggested that the production of water due to the rise of methane in the atmosphere over the last century is what allows these clouds to be observed so frequently now, when they were never reported until the end of the last century. We have also shown that the brightness of the clouds may vary diurnally in response to a diurnal tide, explaining a brightness difference between ground based and satellite observations which were taken at different times of day.

E. Journal Publications: P.Hamill, R.P. Turco, and O. B. Toon, On the Growth of Nitric and Sulfuric Acid Aerosol Particles Under Stratospheric Conditions, J. Atmos. Chem. 7, 287-315 (1988). G.E. Thomas, J.J. Olivero, E. J. Jensen, W. Schroeder, and Owen B. Toon, Relation Between Increasing Methane and the Presence of Ice Clouds at the Mesopause. Nature, 338,490-492,(1988). O.B. Toon, et al., Physical Processes in Polar Stratospheric Ice Clouds. J. Geophys.,in Press (1988).R.P. Turco, O.B. Toon,P. H. Hamill, Heterogeneous Physicochemistry of the Polar Ozone Hole, J. Geophys. Res. in press (1989). S. Kinne, O. B. Toon, G.C. Toon, C.B. Farmer, Measurements of the Size and Composition of Particles in Polar Stratospheric Clouds from Infrared Solar Absorption Spectra, J . Geophys. Res. in press (1989).